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Thermo-reversible silicone elastomer with remotely controlled self-healing

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Abstract

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Nowadays, silicone elastomers are utilized in a wide range of applications, such as artificial muscles in the shape of dielectric elastomers, medical implants, soft robotics, microfluidic devices, and commodity products. Despite their valuable properties, silicone elastomers are due to their covalent nature thermoset polymers, thus not easily recyclable. It is a demanding challenge to turn silicone into thermoplastic, healable, and recyclable materials. For instance, introduction of hydrogen bonds is a useful technique to provide silicone elastomers with thermoplastic properties and self-healing functionalities. Here we present the synthesis of a copolymer via free radical polymerization of monomethacryloxypropyl terminated polydimethylsiloxane (PDMSMA) and 6-methyl-2-ureido-4[1H]-pyrimidone methacrylate (UPyMA). The novel copolymer possesses thermoplastic properties due to the reversible nature of the UPy self-associating dimers.^[1] In addition, self-healing properties were sought by alternative approaches. In particular, induction heating^[2] represents a novel method to externally trigger healing of polymers. This method consists of the incorporation of magnetic particles in the matrix and the exposure of the composite to an alternating magnetic field (AMF). As a consequence, the heat developed by the process activates network rearrangement. Controlled self-healing by use of an AMF generates heat rapidly and locally, and, above all, it is contactless.

In this work, we successfully report the development of a novel thermoplastic and self-healing silicone elastomer, namely P(PDMSMA-co-UPyMA). The remotely controlled healing of the damaged material was performed through exposure to an AMF of the composite elastomer with 20wt% Fe₃O₄ particle filler. Self-healing efficiency was determined by percentage of restored tensile stress and tensile strain of the healed sample compared to the native material. Scanning electron microscopy (SEM) was exploited to evaluate the morphology of samples at the healed interface. Moreover, the cross-linked copolymer was proven remouldable multiple times without showing considerable physical degradation. Hence, this material may be considered as an excellent candidate for recyclable silicone elastomers. In addition, the described self-healing approach was applied using the same conditions to another previously reported copolymer P(MEA-co-UPyMA).^[3] P(PDMSMA-co-UPyMA) and P(MEA-co-UPyMA) bear identical self-complementary motifs, although differing in the main repeating unit. Comparison of their self-healing performance under unchanged conditions aimed at assessing the versatility of the described self-healing method.

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